RESEARCH ARTICLE

Hydrothermal carbonization as an alternative sanitation technology: process optimization and development of low-cost reactor [version 1; peer review: 2 approved with reservations]

Jae Wook Chung [ID]1, Gabriel Gerner1, Ekaterina Ovsyannikova2, Alexander Treichler3, Urs Baier3, Judy Libra [ID]4, Rolf Krebs1

1Institute of Natural Resource Sciences, Zurich University of Applied Sciences, Wädenswil, 8820, Switzerland
2Institute of Agricultural Engineering, University of Hohenheim, Stuttgart, 70599, Germany
3Institute of Chemistry and Biotechnology, Zurich University of Applied Sciences, Wädenswil, 8820, Switzerland
4Postharvest Technology, Leibniz Institute for Agricultural Engineering and Bioeconomy, Potsdam-Bornim, 14469, Germany

Abstract

Background: The provision of safe sanitation services is essential for human well-being and environmental integrity, but it is often lacking in less developed communities with insufficient financial and technical resources. Hydrothermal carbonization (HTC) has been suggested as an alternative sanitation technology, producing value-added products from faecal waste. We evaluated the HTC technology for raw human waste treatment in terms of resource recovery. In addition, we constructed and tested a low-cost HTC reactor for its technical feasibility.

Methods: Raw human faeces were hydrothermally treated in a mild severity range ($\leq$ 200 °C and $\leq$ 1 hr). The total energy recovery was analysed from the energy input, higher heating value (HHV) of hydrochar and biomethane potential of process water. The nutrient contents were recovered through struvite precipitation employing process water and acid leachate from hydrochar ash. A bench-scale low-cost reactor (BLR) was developed using widely available materials and tested for human faeces treatment.

Results: The hydrochar had HHVs (23.2 - 25.2 MJ/kg) comparable to bituminous coal. The calorific value of hydrochar accounted for more than 90% of the total energy recovery. Around 78% of phosphorus in feedstock was retained in hydrochar ash, while 15% was in process water. 72% of the initial phosphorus can be recovered as struvite when deficient Mg and NH_4 are supplemented. The experiments with BLR showed stable operation for faecal waste treatment with an energy efficiency comparable to a commercial reactor system.

Open Peer Review

Approval Status ✓ ?

	1	2
version 2 (revision) 10 Jun 2022	✓ view	
version 1 26 Nov 2021	? view	? view

1. **Luca Fiori** [ID], University of Trento, Trento, Italy

   Donato Scrinzi [ID], University of Trento, Trento, Italy

2. **Amit Gross** [ID], Ben-Gurion University of the Negev, Be'er Sheva, Israel

Any reports and responses or comments on the article can be found at the end of the article.

Conclusions: This research presents a proof of concept for the hydrothermal treatment of faecal waste as an alternative sanitation technology, by providing a quantitative evaluation of the resource recovery of energy and nutrients. The experiments with the BLR demonstrate the technical feasibility of the low-cost reactor and support its further development on a larger scale to reach practical implementation.

Keywords
hydrothermal carbonization, sanitation, energy, nutrient, faecal sludge, resource recovery, struvite, fertilizer

This article is included in the Excellent Science gateway.

This article is included in the Green Chemistry collection.

Corresponding author: Jae Wook Chung (shoutjx@gmail.com)

Author roles: Chung JW: Conceptualization, Funding Acquisition, Investigation, Writing – Original Draft Preparation; **Gerner G**: Conceptualization, Funding Acquisition, Project Administration; **Ovsyannikova E**: Investigation; **Treichler A**: Investigation; **Baier U**: Conceptualization, Supervision; **Libra J**: Conceptualization, Funding Acquisition, Supervision; **Krebs R**: Conceptualization, Funding Acquisition, Supervision

How to cite this article: Chung JW, Gerner G, Ovsyannikova E *et al.* **Hydrothermal carbonization as an alternative sanitation technology: process optimization and development of low-cost reactor [version 1; peer review: 2 approved with reservations]** Open Research Europe 2021, **1**:139 https://doi.org/10.12688/openreseurope.14306.1

First published: 26 Nov 2021, **1**:139 https://doi.org/10.12688/openreseurope.14306.1

Plain language summary

In less developed places where modern wastewater treatment systems are unavailable, relatively simple onsite technologies are implemented for human waste treatment. Faecal sludge produced from those facilities needs proper treatment to ensure public health and resource recovery. In this research, we studied raw human faeces treatment with hydrothermal carbonization technology, which converts wet biomass into a charcoal-like solid material (hydrochar) and organic-rich liquid (process water) at relatively mild temperature (~200 °C) under pressurized conditions (~20 bar). We tested several reaction parameters to find an optimum point for the best energy recovery. The total energy value of the resulting materials was analysed by measuring the heating value of hydrochar and biogas production from process water. The remaining nutrient contents in hydrochar ash and process water was recovered through struvite precipitation which can be used as a slow-releasing fertiliser. Hydrochar derived from human faeces showed comparable heating values to bituminous coal. Around 70 % of initial phosphorus in faeces was recovered as struvite with supplements of magnesium and ammonium from external sources. Also, we developed a bench-scale low-cost reactor using ordinary pipe-fitting materials and tested it for faecal waste treatment. These results present the initial evidence for technical feasibility and resource recovery. Further research for testing various faecal sludges in field conditions and developing pilot-scale low-cost reactors are encouraged.

Introduction

Untreated human excreta pose serious risks to public health and environment. In 2020 almost half of the global population still does not have access to safely managed sanitation services that properly contain and treat the faecal waste. In the least developed countries (LDCs), the conditions are most severe with only 26% of people covered by suitable services. To be able to achieve the Sustainable Development Goal (SDG) aimed at universal access to safely managed sanitation services by 2030, the current rate of progress in LDCs must be accelerated 15 times (WHO & UNICEF, 2021).

In the places where centralised sewer collection and wastewater treatment systems are unaffordable or inefficient, onsite or decentralised technologies such as pit latrines and septic tanks can be implemented as the collection method. The faecal sludge generated from these technologies requires appropriate service chains that target the safe end-use or disposal of the final product (Strande *et al.*, 2014). Since one of the main reasons for insufficient service provision is the high economic burden placed on stakeholders, the development of profitable business opportunities could lead to breakthroughs in this sector (Boot & Scott, 2009). Although various options for final product valorisation have been proposed, including fuel briquette, biogas generation and protein feed (Black soldier fly larvae), the most common practice being implemented is agricultural application through composting which also has the lowest economic value of the options (Mallory *et al.*, 2020).

In recent years, hydrothermal carbonization (HTC) has been investigated to treat faecal waste for sanitisation and resource recovery. It is a thermochemical process that converts wet biomass into more carbonaceous materials at relatively low temperatures (~200°C) under pressurized conditions (~20 bar) which results from autogenic pressure development. Since HTC treats wet feedstocks, it has the advantage of lower energy requirements compared to conventional dry pyrolysis technologies that need coupling with an energy-intensive drying process (Libra *et al.*, 2011). The output from the HTC treatment usually has the form of a thick slurry which further can be separated into solid hydrochar (HC) and organic-rich process water (PW). These products have a wide range of potential applications e.g. as energy sources, soil amendment, fertiliser, and adsorbent (Sharma *et al.*, 2020). Hydrothermal treatment of different types of faecal wastes has produced HC materials with enhanced higher heating values (HHV) ranging from 16 – 28 MJ / kg, using primary sewage sludge (Danso-Boateng *et al.*, 2013), raw human faeces (Wüst *et al.*, 2019; Yahav Spitzer *et al.*, 2018), faecal sludge (Fakkaew *et al.*, 2018) and their simulants (Afolabi *et al.*, 2017). In general, applying higher temperatures and longer retention times has been found to increase the HHV in HC materials but with a lower solid yield. In addition to improving the fuel value of faecal waste, nutrients can be recovered from the HTC products. Wüst *et al.* (2019) reported that a considerable proportion of phosphorus and nitrogen in raw human waste was retained in the solid phase after the HTC treatment, while alkali salts were dissolved in PW. Especially the recovery of phosphorus, an essential nutrient for living organisms, can be an important advantage in LDC sanitation services, since it is a limited resource globally (Cordell *et al.*, 2009; Cordell *et al.*, 2011) and in many local agricultural applications. Diminishing high-quality phosphorus sources and the exploitation of low-quality sources with higher transport, extraction and processing costs cause a greater cost burden on farmers (Desmidt *et al.*, 2015). The inclusion of a technology in the local sanitation service chain that can aid in the reutilisation of phosphorus in waste streams for local food production may lead to the required breakthroughs in this sector.

Despite the strong potential of HTC as a sanitation technology, there is still not enough research pursuing its implementation in less developed communities with limited technical and economic resources. Since the major cost factors of HTC technology arise from reactor construction and energy consumption for treatment, optimizing operational parameters in a low-temperature range would be beneficial in lowering both capital and operating costs (Song *et al.*, 2019). Previous research has highlighted the potential for cost reduction in HTC reactor construction. A batch type reactor with 20 L capacity was constructed employing a relatively simple design and function at a material cost of around CHF 11K (excluding personnel and certification cost), which was several times cheaper than a commercial reactor system with comparable treatment capacity (Robbiani, 2013).

The purpose of this research was to investigate HTC technology as an alternative sanitation technology for faecal waste treatment, mainly focussing on the technical feasibility of the low-cost approach and its potential for resource recovery of energy

and nutrients. Firstly, raw human waste was treated in a HTC commercial reactor at a variety of mild operational conditions. The total energy recovery was evaluated based on the energy input to the process and recovery through the products of both phases. The resulting materials from the best case were analysed for phosphorus recovery through the struvite precipitation process. Secondly, a low-cost reactor employing ordinary pipe-fitting materials and a simple heating method was constructed and validated for faecal waste treatment using the selected operational conditions. Based on these results, a scenario for energy and nutrient recovery through hydrothermal treatment of faecal waste is discussed.

Methods

All experimental data was processed by Microsoft Excel (2018).

Human waste feedstock and hydrochar

Two stocks of human faeces were collected at a composting toilet company (Kompotoi, Zurich, Switzerland) and processed in separate batches. Ethical approval was not required for this study because the materials were collected from toilets used by anonymous users. First the larger additives, toilet paper and woody cover materials, were sorted out from the raw stock. The collected material was then autoclaved at 121 °C for 20 min and dried in an oven at 105 °C overnight. The remaining smaller additives other than faecal mass was removed from the dry stocks, before being ground in a mechanical grinder (GM 300, Retsch, Haan, Germany) and stored at 4 °C until used. The first (Faeces-1) and the second (Faeces-2) stocks were used for optimization experiments and BLR experiments, respectively. The feedstock for each HTC run was prepared by mixing the designed amount of dry faeces with demineralised water to a solids content of 20% dry matter (DM). Then, this mixture was placed at room temperature overnight to ensure sufficient reassociation of dry faeces and water. After the HTC, the resulting slurry from the HTC experiment was separated by either vacuum filtration employing filter paper with a pore size of 11 μm (Whatman, Maidstone, UK) or centrifugation (5910R, Eppendorf, Hamburg, Germany) at 4300 RCF for 3 min. The HC was dried in an oven at 105 °C overnight. The resulting HC and PW samples were stored at 4 °C until used. Solid samples for chemical analyses were further powdered by a ball mill (MM400, Retsch GmbH, Haan, Germany).

HTC procedure and reactors

Two sets of HTC experiments for faecal waste treatment were carried out. Firstly, selected operational parameters were tested with a commercial reactor system to find the condition for best energy recovery ratio (energy content / energy input). Then, a bench-scale low-cost reactor (BLR) was developed and tested for HTC conditions obtained from the previous optimization experiments.

Process optimization in commercial batch reactor. In the optimization study, several process parameters were tested to find an optimum point for best energy recovery ratio using a commercial batch reactor system (Kiloclave, Büchi AG, Uster, Switzerland). Briefly, 480g feedstock (Faeces-1) was

hydrothermally treated in a pressure vessel with an active volume around 640 mL (vessel volume subtracted by the volume of in-vessel equipment e.g. stirring rod, baffle, and stem of temperature probe) at an approximate fill-up rate of 3 / 4. The reactor jacket was equipped with a temperature sensor and an electric heating mantle of a 1000 W capacity. The temperature (TR) and pressure inside the reactor were monitored in a 20-seconds interval. The heating intensity was regulated by a proportional–integral–derivative (PID) controller as programmed for each test condition. The energy input for the reaction was estimated by an integration of the duration-throttle of the heating mantle. To approximate the conditions expected in low-cost reactor systems, stirring and active cooling after the reaction were not applied. Two operational procedures were followed: 1) the reactor was heated to the target temperature and held there for a holding time (10 or 60 minutes), or 2) the heating was stopped immediately when the target temperature was reached, called Touch & Down (TD). This new operational regime was tested to simulate the conditions where automated temperature control is unavailable.

In total, seven experimental conditions were tested in duplicate (Table 1) for HTC process optimization. The first two digits of the Test ID indicate the reactor temperature and the third digit represents the holding time at the temperature. e.g. Test-161 for HTC at 160 °C for 10 minutes and Test-206 for HTC at 200 °C for 60 minutes. The TD case, where the heating stopped when TR = 200 °C, is named Test-200.

The reaction severity factor (SF) of each condition was presented according to the Equation (1) and Equation (2) (Heidari *et al.*, 2019). Also, an adjusted severity factor (SF_{adj}) was obtained by substituting t with t_{adj} in the Equation (1) for better comparison of TD cases with other tests.

$$R = t\,e^{\frac{(T-100)}{14.75}} \qquad (1)$$

$$SF = \log R \qquad (2)$$

where R is the reaction ordinate (min); t is the reaction time (min); T is the reaction temperature (°C); t_{adj} is the duration at reaction temperature ± 3 % (min).

Bench-scale low-cost reactor. The BLR was constructed by a pressure vessel manufacturing company (Bero Technik, Sirnach, Switzerland). Details of the designing consideration, technical drawing and parts information are available as *Extended data* (Chung, 2021). Briefly, a 2 L vessel was fabricated using austenitic stainless steel pipe and fitting materials (EU code - 1.4404 / US - 316L) through circumferential welding technique. The main reactor body consists of a blind flange, weld-neck flange, pipe and end cap. It was equipped with a bimetal temperature gauge, a bourdon tube pressure gauge and a bleed valve. For safety assurance, an overpressure valve and a rupture disc were installed. The BLR was mounted on a heating unit obtained / modified from previous research (Bleuler *et al.*, 2021). A portable camping stove with a 3000 W capacity (Easy Fuel Duo, Primus, Stockholm,

Table 1. Features of hydrothermal carbonization tests and characteristics of resulting materials. Results from hydrothermal carbonization experiments.

†‡Test ID	Temp. (°C)	Time (min)	*Severity Factor (SF)	Time at Target Temp. ± 3 % (min)	**Adj. SF	Peak Pressure (bar)	pH	C	H	N	#S	§O	Ash	Hydrochar Yield (%)	Process Water (g)
Faeces-1								49.6	6.7	4.2	0.5	27.2	10.2		
Faeces-2								48.8	7.4	4.5		28.4	10.9		
161	160	10	2.77	15.9 ± 0.6	2.97	5.9	5.7	51.6	7.4	3.6	0.4	27.5	9.4	76.0 ± 0.7	270.1 ± 1.3
166	160	60	3.54	72.5 ± 3.5	3.63	5.9	5.5	52.6	7.5	3.4	0.5	26.3	9.9	71.3 ± 0.5	261.7 ± 5.4
181	180	10	3.36	17 ± 1.0	3.58	10.1	5.5	52.9	7.5	3.4	0.4	26.0	9.9	70.7 ± 0.7	272.5 ± 8.6
186	180	60	4.13	71.9 ± 0.9	4.21	10.7	5.2	54.7	7.6	3.2	0.4	23.9	10.3	67.7 ± 0.3	279.1 ± 3.6
200	200	1 s	0.02	4.3	3.58	16.1	5.3	54.5	7.7	3.2	0.5	23.9	10.2	69.9 ± 0.2	284.0 ± 5.0
201	200	10	3.94	20.9 ± 0.2	4.26	16.7	5.2	55.3	7.6	3.2	0.4	23.5	11.0	67.6 ± 0.1	282.7 ± 5.1
206	200	60	4.72	73 ± 2.3	4.80	19.8	5.2	56.4	7.6	3.1	0.4	21.7	10.8	65.7 ± 0.7	279.7 ± 13.1
BLR-161	160	10	2.77	24	3.14	7.3	5.9	52.2	7.6	3.8		25.4	11.0	70.7	883.0
BLR-200	200	1 s	0.02	12	4.02	21.0	5.7	55.0	7.5	3.7		21.8	12.1	65.5	890.8

† Tests performed with the commercial reactor system: n=2, and results from composite samples prepared by combining resulting materials from both runs. The values indicate the average of two observations ± difference to individual data points.

‡ Tests performed with the BLR: n=1.

* Severity factor, Heidari *et al.,* 2019.

** Adjusted severity factor based on the duration at set temperature ± 3 %.

Sulfur contents obtained from ICP-OES analysis

§ Oxygen contents estimated by subtracting the CHNS and ash fraction from 100 %.

Sweden) was used to heat the BLR. The weight of the gas bottle was measured before and after the HTC to estimate energy input.

Two HTC conditions chosen from the optimization experiments were performed in the BLR in singlicate, Test-161 and Test-200. Hereafter, these tests were noted as BLR-161 and BLR-200, respectively. For each run, 1.5 kg of faecal feedstock (Faeces-2) with a solids content of 20% DM filled approximately 3 / 4 of the reactor volume. TR and pressure inside the reactor were monitored by gauge reading. The surface temperatures of the blind flange and gauge heads were measured using an infrared temperature gun (Fluke, Everett, US) with black insulation tape applied to the measurement surfaces. The pressure and temperature values were monitored in 3 min intervals. The heating intensity was controlled manually by adjusting the valve to the gas stove.

Material characterization
The pH of the resulting slurry from HTC was measured with a pH meter (HQ40d, Hach, Loveland, US). Ash contents of HC and faeces samples were obtained according to DIN EN 14775, using a muffle furnace (L 40 / 11 BO, Nabertherm, GmbH, Lilienthal, Germany). The resulting HC ash was used for subsequent experiments for nutrient recovery. Volatile solid (VS) of PW samples were measured according to 2540-E. CHN compositions of solid samples were measured by a CHN analyser (TruSpec CHN, Leco, St. Joseph, US). Elemental analysis for PW, HC and HC ash was performed by the ICP-OES technique (Agilent 715, Agilent, Santa Clara, US) as previously reported (Ovsyannikova *et al.,* 2020). NH_4-N concentration of PW was measured by the Hach-Lange cuvette test (LCK 304, Hach, Loveland, US).

Energy analysis
The HHV of HC and dry faeces were measured with a bomb calorimeter (C200, IKA, Staufen, Germany). Biomethane potential (BMP) of the PW samples were analysed in batch tests according to VDI-4630 (VDI, 2006) using an AMPTS system (BPC Instruments, Lund, Sweden). Briefly, microbial inoculum (anaerobic sludge) was collected at a wastewater treatment plant (ARA Rietliau, Wädenswil, Switzerland) and stabilised

at room temperature for three days. 500 mL Schott bottles were filled with 390 g of inoculum and 10 g of the PW (substrate). These bottles were purged with nitrogen gas for 3 min and placed in water baths set at 37°C. The gas generated from bottles passed through 3 M NaOH solution supplemented with thymolphthalein (pH indicator) to remove CO_2 from the raw biogas, and the rest was considered CH_4. Whole experimental procedures were validated by testing positive control (seeded with standard cellulose substrate) and blank (inoculum-only) samples. All substrates and blank tests were triplicated, and the average values were used for further calculation.

The process energy input for the experiments with BLR was obtained according to the model previously suggested by Danso-Boateng et al. (2015) with minor modifications as follows (3).

$$(\text{Energy input}) = (\text{Energy to heat reactor}) + (\text{Energy to heat dry faeces})$$
$$+ (\text{Energy to heat water}) + (\text{Heat of reaction}) + (\text{Heat loss}) \quad (3)$$

$$[(M_{g0} - M_g) * HV] = [M_r * C_{pr} * (T - T_0)] + [M_f * C_{pf} * (T - T_0)]$$
$$+ [M_w * (H_r - H_{T0})] + [M_0 * \Delta HR] + (\text{Heat loss}) \quad (4)$$

where M_{g0} is the initial mass of gas bottle (kg); M_g is the final mass of gas bottle after HTC run (kg); HV is the heating value of gas, estimated from its composition (49.4 MJ kg^{-1}); M_r is the reactor mass measured (21.5 kg); C_{pr} is the specific heat capacity of reactor material (0.5 kJ kg^{-1} K^{-1}); T_0 and T are the initial temperature and the target temperature (K), respectively; $M_f = M_0$ is the mass of dry faeces in the feeding slurry (kg); C_{pf} is the specific heat capacity of dry sewage sludge as a simulant for faeces (1.7 kJ kg^{-1} K^{-1}) (Namioka et al., 2008) ; M_w is the mass of water in feeding slurry; H_{T0} is the enthalpy of water at T_0; H_T is the enthalpy of saturated water at T; ΔHR is the heat of reaction reported for 4 h reaction of faecal sludge (- 0.20 MJ kg^{-1} at 160 °C and - 0.70 MJ kg^{-1} at 200 °C) (Danso-Boateng et al., 2015). The heat loss was calculated by difference. Finally, the energy recovery ratio for the HTC process is defined as the ratio of energy content to energy input. The energy content of HC and PW was considered.

Nutrient recovery through struvite precipitation

For the HTC condition yielded the best energy balance in optimization experiments (Test-161), recovery of phosphorus from hydrochar ash through struvite precipitation was investigated. Experimental procedures of previous research (Ovsyannikova et al., 2020; Ovsyannikova et al., 2021) were adopted with minor modifications. To extract the phosphorus content from ash, 0.9 g of HC ash was added to 10 mL 1M H_2SO_4 to have the molar ratio between H^+ and P to 4:1 (Petzet et al., 2012) and stirred overnight. The struvite precipitation was performed using 2 mL of the leachate with the supplement of PW (as NH_4-N source) and $MgCl_2.6H_2O$ (as Mg source) at designed doses for establishing the same molar ratio among P, NH_4-N and Mg in the precipitation batch. For struvite precipitation, the pH of the solution was adjusted to 8 by the addition of 1M NaOH. The solution was stirred at 500 rpm during the pH setting and reaction time of 45 min.

The precipitate formed was recovered by filtration through the filter paper (5–13 μm, VWR, Ulm, Germany) and dried at 35°C. The ICP-OES and CHN analyses were performed to investigate its composition. Struvite structure in the precipitate was verified by XRD technique using a D8 Advance (Bruker, Billerica, US) employing Cu radiation through Ni-filter. The $K_{\alpha2}$ component was numerically subtracted. The diffraction pattern was identified with the database of International Centre for Diffraction Data (ICDD).

Results

HTC of faeces

All HTC treatments in this study showed distinctive transformation of the faeces when compared to the untreated raw material (Chung et al., 2021). The odour associated with faeces was replaced with a smell like spent coffee grounds. The brown colour of feedstocks changed into dark brown-black in the end products. All resulting slurries except for the ones from Test-161 had good dewaterability and were vacuum-filtered through 11 μm pores. Due to its inferior dewaterability, the slurry from Test-161 was separated into solid and liquid phases by centrifugation. Table 1 summarizes the general features of HTC experiments and results of ultimate analyses of HC samples. The HC yields and changes in the major elemental composition of HCs showed dependency on the calculated SF values. Less HC was recovered from experiments with higher SFs, while the HC obtained at higher SFs had improved characteristics as solid fuel, i.e. they had higher carbon and lower oxygen contents. The adjusted SF was seen to be a more reliable indicator of the reaction characteristics when comparing TD cases with limited holding time at the target temperature, i.e. Test-200 and BLR-200, to non-TD cases.

The peak pressure during HTC was primarily determined by the water vapor saturation pressure (Psat) which increased with TR. Substantial pressure development over Psat occurred for longer holding times and higher temperatures. Figure 1 shows the TR and pressure development from a few selected tests. The pressure increase over the holding time is visible when comparing the TD run with 60-minute holding, Test-200 vs. 206 in the commercial reactor (Figure 1a). After reaching the target temperature of 200 °C, the pressure increased continuously above Psat as heating was applied to maintain the TR at the target temperature. The same phenomenon, but to a lower extent, was observed in the comparison between Test-181 and 186, while Test-161 and 166 did not show a significant difference in their peak pressure. This indicates that the hydrothermal conversion of the feedstock is taking place with gas generation during the holding time. Compared to the commercial reactor, the BLR had a lower heating rate and took almost twice as long to reach the target temperature 200 °C (Figure 1b). Due to this slower rate, the TD run in the BLR (BLR-200) remained at temperatures above 180 °C much longer than the TD run in the commercial reactor (Test-200). Much more pressure was produced in BLR-200 than in Test-200. This resulted in a higher peak pressure than the one obtained from the same test condition performed with the commercial reactor. Also, it was noteworthy in terms of safety assurance that the pressure development around the target temperature of 200°C
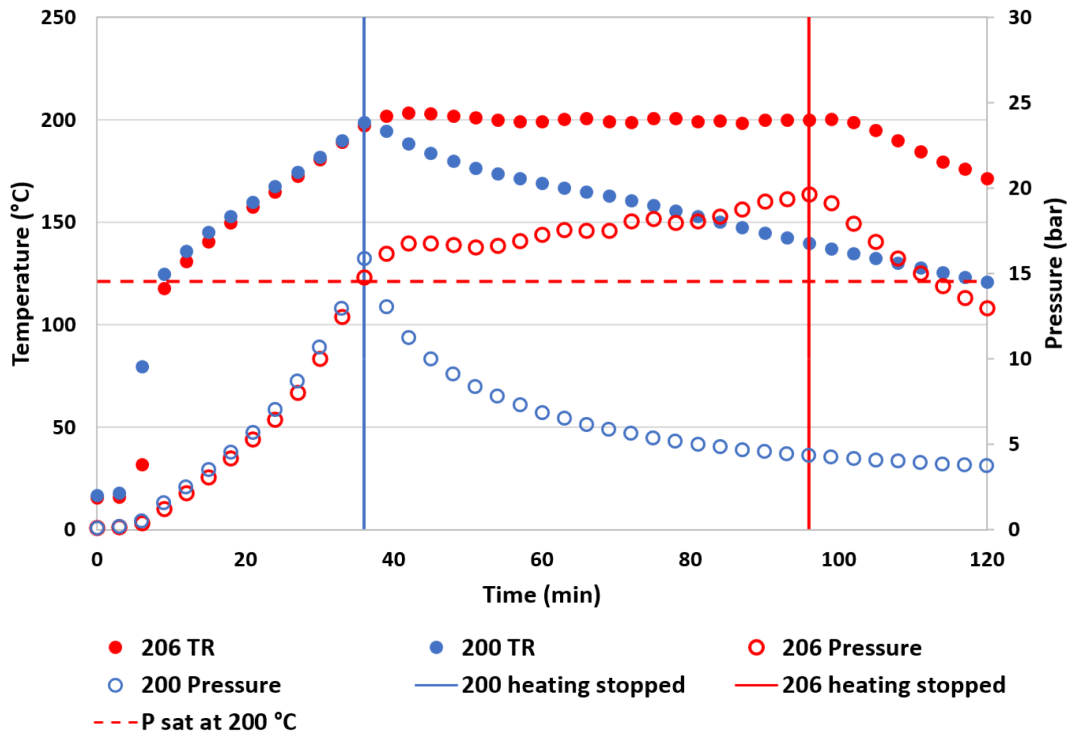
(a)
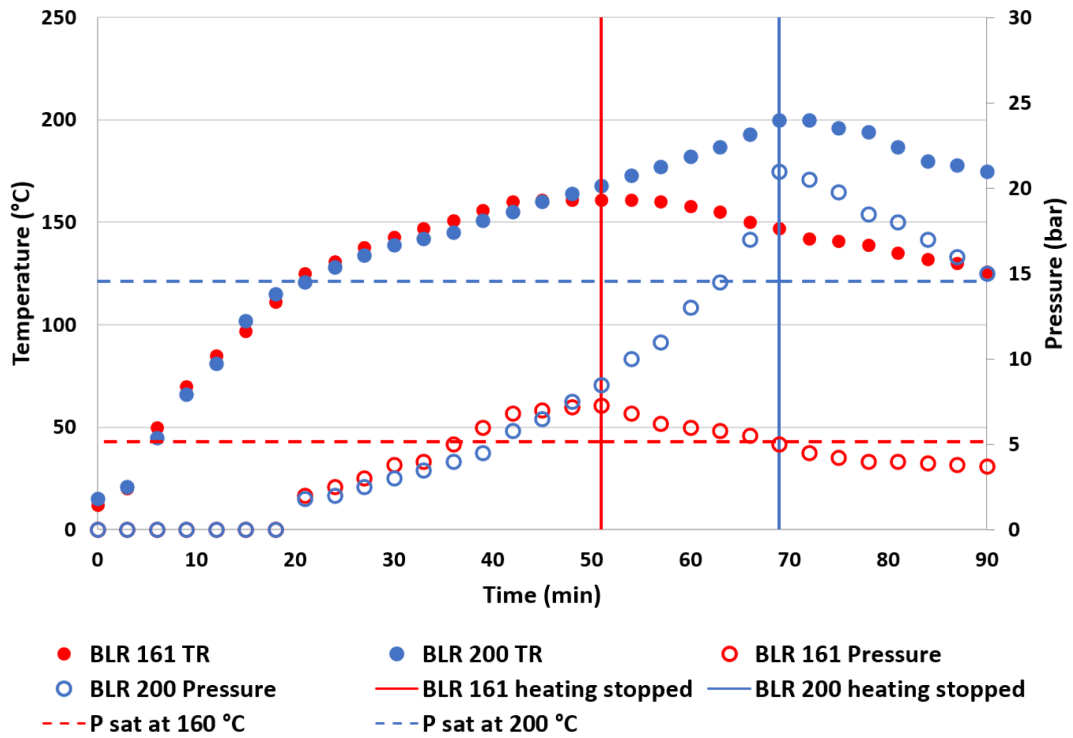
(b)

Figure 1. Temperature and pressure development during hydrothermal carbonization (HTC). (**a**) Test-200 and 206 performed with the commercial reactor, (**b**) Test-200 and 161 performed with the bench-scale low-cost reactor.

in BLR was vigorous and approached the maximum operational pressure. In all cases, the pressure inside the reactor started to decrease immediately when the heating stopped, even while the TR was still greater than 180°C. Throughout all experiments, the surface temperature of pressure and temperature gauge heads installed at BLR were below their allowable ambient temperature limit of 60°C and 70°C, respectively.

Energy contents

The energy input for the process test conditions and energy content of the resulting materials are presented in Table 2. The combination of these two values in the energy recovery ratio (energy content / energy input) provides an indicator to compare the overall energy efficiency for the process and products of both phases. Looking first at HC; the HTC experiments with higher reaction severity densified the energy content in the solid phase to a greater extent, resulting in HHV values ranging between 23.2 and 25.2 KJ/g. In contrast, as seen in Table 1, the trend for the HC yield is inversely related to SF_{adj}. Therefore, despite the differences in HHV, the energy content of HC was similar for all test conditions, ranging from 1.59 to 1.69 MJ. For PW, the energy content was determined in BMP experiments and also varies only slightly,

between 0.08 and 0.13 MJ. In all cases, more than 90 % of the biogas was generated within the first five days (data not shown), showing good potential for recovery. However, only 4 – 7 % of the total energy content was recovered from the PW; the majority of the energy was recovered in the HC. Because the energy content from HC and PW were comparable for all test conditions employed in this research, the energy recovery ratio was primarily determined by the energy input. Since more energy input is required at the higher SF_{adj} conditions, this leads to lower energy recovery ratios as temperature and holding time are increased. While the Test-161 yielded more energy than what was invested, the Test-206 recovered only 52.4 % of its energy input.

The experiments performed with BLR showed a better energy efficiency (i.e. energy consumption for carrying out the same task) than the commercial reactor employed in optimization experiments. To treat 1 kg dry faeces, the commercial reactor required 16.6 MJ for Test-161 and 23.0 MJ for Test-200. The values from BLR for the same test condition were 16.0 MJ for BLR-161 and 20.5 MJ for BLR-200. The energy input for two tests performed with the BLR showed a similar distribution between the energy uses (Table 3). About half of

Table 2. Energy input for test conditions and recovery from resulting materials. HTC=hydrothermal carbonization.

Test ID	Energy input during HTC (MJ)	Hydrochar				Process water					† Total energy recovery ratio
		Weight (g)	HHV (KJ/g)	Energy content (MJ)	Ratio to E. input (%)	Weight (g)	VS (g / g PW)	* CH_4 yield (g CH_4 / g VS)	Energy content (MJ)	Ratio to E. input (%)	
161	1.59 ± 0.02	72.9 ± 0.7	23.2	1.69 ± 0.02	106.4	270.1 ± 1.3	0.051	0.13 ± 0.04	0.09	5.5	111.9
166	2.35 ± 0.05	71.3 ± 0.5	23.7	1.69 ± 0.01	71.9	261.7 ± 5.4	0.054	0.11 ± 0.01	0.08	3.4	75.4
181	2.10 ± 0.03	67.7 ± 0.8	23.7	1.61 ± 0.02	76.7	272.5 ± 8.6	0.057	0.13 ± 0.01	0.10	4.7	81.4
186	2.81 ± 0.11	64.9 ± 0.3	24.6	1.60 ± 0.01	57.1	279.1 ± 3.6	0.054	0.15 ± 0.00	0.11	4.0	61.1
200	2.21 ± 0.09	67.0 ± 0.2	24.5	1.65 ± 0.00	74.6	284.0 ± 5.0	0.056	0.15 ± 0.03	0.13	5.8	80.4
201	2.41 ± 0.05	64.8 ± 0.0	25.0	1.62 ± 0.00	67.2	282.7 ± 5.1	0.052	0.18 ± 0.05	0.10	4.2	71.4
206	3.28 ± 0.05	63.1 ± 0.7	25.2	1.59 ± 0.02	48.7	279.7 ± 13.1	0.047	0.16 ± 0.03	0.12	3.7	52.4
BLR-161	4.80	212.0	23.2	4.93	102.6	883.0	0.048	not available			102.6 +
BLR-200	6.14	196.6	24.4	4.80	78.2	890.8	0.047				78.2 +
Faeces-1	96 g in feedstock		22.8	2.19		not applicable					
Faeces-2	300 g in feedstock		21.4	6.42							

- Average value ± difference to individual data points n=2 or * standard deviation n=3.

- † (energy content of HC + PW) / energy input.

the energy was used to heat the reactor and feedstock, and the other half was lost during the treatment as heat loss. The exothermic energy from the hydrothermal reaction of feedstock contributed to the total energy demand to a limited extent (< 4% of energy input).

Nutrient contents

The overall inorganic content of the solids as indicated by the ash content was hardly changed by the HTC treatment. The

range remained similar to that of the dry faeces, 9.4–11.0 wt % (Table 1). However, the trends of the individual nutrients in the HC differed: the Ca and P concentrations increased with reaction severity, while the S, K, Mg and Na concentrations in the HCs decreased or remained comparable to the dry faeces. Measurement of the nutrients in the ash after HC combustion showed a 10-fold increase in concentrations for almost all elements except for sulfur. During the HTC treatment, a considerable amount of nutrients was released to the liquid phase (PW). While the concentration of most nutrients in PW was similar for all test conditions, the NH_4-N concentration showed a trend that higher reaction severity resulted in higher NH_4-N values.

Table 4 summarizes the partitioning and fate of P and Mg through the hydrothermal treatment and subsequent HC combustion. These are of interest because they can be recovered further through struvite precipitation. Despite some differences in the concentration between test conditions (Table 5), the total mass of each nutrient contained in HC was comparable for all tests. The increase in P concentration with higher reaction severity was compensated by the higher mass loss and lower HC yields, so that the mass of P in HC remained the same. Of the total mass in the feedstock, 75.6 - 81.0 % P remained in HC,

Table 3. Energy input for bench-scale low-cost reactor operation.

Contents	BLR-161	BLR-200
	MJ (% of energy input)	
Energy input	4.80 (100.0)	6.14 (100.0)
Energy to heat reactor	- 1.59 (33.1)	- 1.99 (32.4)
Energy to heat faeces	- 0.08 (1.6)	- 0.09 (1.5)
Energy to heat water	- 0.75 (15.6)	- 0.95 (15.4)
Heat of reaction	0.06 (1.3)	0.21 (3.4)
Heat loss	- 2.44 (50.9)	- 3.32 (54.1)

Table 4. Phosphorus and magnesium balance through hydrothermal carbonization and subsequent combustion of hydrochar. PW=process water; HTC=hydrothermal carbonization.

Test ID	Total amount in HC (mg)	Partitioning ratio in HC (%)	Total amount in PW (mg)	Partitioning ratio in PW (%)	Loss from HTC (%)	Total amount in Ash (mg)	Percentage from HC found in Ash (%)	Loss from HTC + Combustion (%)
				Phosphorus				
161	1084.9	79.0	205.3	14.9	6.1	1080.3	99.6	6.4
166	1089.9	79.4	197.4	14.4	6.2	1079.0	> 99.9	7.0
181	1069.6	77.9	211.7	15.4	6.7	1008.4	94.3	11.1
186	1038.2	75.6	232.9	17.0	7.4	1027.8	> 99.9	8.2
200	1048.0	76.3	230.9	16.8	6.9	1027.0	98.0	8.4
201	1077.5	78.5	248.3	18.1	3.4	1006.4	93.4	8.6
206	1112.2	81.0	189.9	13.8	5.2	1032.5	92.8	11.0
Faeces-1	feedstock 1373.2				Not applicable			
				Magnesium				
161	345.2	61.5	194.1	34.6	3.9	342.3	99.2	4.5
166	335.1	59.7	195.1	34.7	5.6	322.0	96.1	7.9
181	312.9	55.7	206.6	36.8	7.5	281.5	90.0	13.1
186	295.2	52.6	224.7	40.0	7.4	285.3	96.6	9.2
200	304.0	54.2	224.8	40.0	5.8	299.1	98.4	6.7
201	297.9	53.1	235.8	42.0	4.9	271.0	91.0	9.7
206	320.8	57.1	215.3	38.3	4.6	289.7	90.3	10.1
Faeces-1	feedstock 561.5				Not applicable			

- Average values of duplicates

Table 5. Nutrient contents in the feedstock, hydrochar, process water and hydrochar-ash samples.

Sample	S	Ca	K	Mg	Na	P	NH$_4$-N[†]
HC				mg / g			
161	4.4 ± 0.1	24.7	5.2 ± 0.3	4.7	1.0	14.9 ± 0.2	
166	4.5 ±0.2	25.6	5.7 ± 0.1	4.7 ± 0.1	1.1	15.3 ± 0.2	
181	4.2 ±0.1	27.4	5.1 ± 0.2	4.6	1.0	15.8 ± 0.1	
186	4.4	27.4	5.3 ± 0.1	4.5	1.0	16.0	
200	4.5 ± 0.2	26.9	5.2 ± 0.1	4.5	1.0	15.6 ± 0.1	
201	4.4 ± 0.1	28.5	5.4 ± 0.1	4.6	1.1	16.6	
206	4.4 ± 0.1	29.1	5.9 ± 0.2	5.1	1.2	17.6 ± 0.1	
Faeces-1	4.9 ± 0.2	20.8 ± 0.4	10.7 ± 0.6	5.8	1.9 ± 0.1	14.3 ± 0.2	not available
Ash				mg / g			
161	13.7 ± 0.3	260.1 ± 1.7	52.0 ± 0.5	49.7 ± 0.7	12.4 ± 0.2	156.9 ± 2.1	
166	12.0 ± 0.2	255.1 ± 2.9	51.0 ± 0.3	46.7 ± 0.6	11.6 ± 0.1	159.1 ± 1.8	
181	12.1 ± 0.1	254.4 ± 2.2	45.3 ± 0.3	42.6 ± 0.4	9.8 ± 0.1	152.6 ± 1.3	
186	12.9	261.9 ± 2.8	47.4 ± 0.6	43.2 ± 0.1	10.6 ± 0.2	157.6 ± 0.7	
200	13.7 ± 0.1	252.0 ± 2.5	49.3 ± 0.8	43.6 ± 0.3	11.2 ± 0.2	149.8 ± 0.7	
201	14.4 ± 0.2	259.2 ± 1.9	45.4 ± 0.8	41.7 ± 0.6	10.5 ± 0.1	155.0 ± 1.7	
206	13.6 ± 0.2	245.5 ± 1.0	45.7 ± 0.6	42.3 ± 0.1	10.8 ± 0.1	150.6 ± 0.5	
PW				mg / L			
161	373.1 ± 12.4	73.6 ± 2.2	2148.5 ± 64.4	733.0 ± 25.9	449.0 ± 12.6	775.2 ± 29.7	899.0
166	388.5 ± 6.5	59.2 ± 1.4	2106.0 ± 40.7	760.4 ± 11.5	433.3 ± 11.2	769.4 ± 13.5	604.4
181	384.6 ± 11.6	93.8 ± 3.3	2081.4 ± 99.2	773.4 ± 21.1	435.8 ± 23.6	792.4 ± 26.9	1033.0
186	403.6 ± 2.8	124.4 ± 1.9	2200.8 ± 46.9	821.3 ± 8.1	447.3 ± 3.7	851.1 ± 10.6	1265.3
200	412.5 ± 3.9	112.4 ± 0.8	2243.9 ± 63.9	807.6 ± 2.7	444.1 ± 3.6	829.5 ± 4.3	1257.3
201	406.9 ± 8.1	191.0 ± 0.8	2262.2 ± 6.0	850.8 ± 10.4	457.7 ± 1.7	895.9 ± 11.6	1367.6
206	399.8 ± 2.4	79.3 ± 0.8	2216.5 ± 41.4	785.2 ± 6.7	456.3 ± 5.4	692.8 ± 8.1	1571.5

- Average value ± standard deviations n=3, noted when it is same or greater than 0.1.

[†] Average values of duplicates

while 13.8 - 18.1 % was found in PW. In contrast, Mg showed a relatively large partitioning ratio into the PW, ranging from 34.6 to 42.0 %, while that for HC ranged from 52.6 to 61.5 %. After the HC was combusted, both P and Mg remained in the ash, with more than 90 % of the total amount recovered. In summary, the partitioning of nutrients was not dependent on the reaction severity of the test conditions employed in this study.

Struvite precipitation
The HC ash from Test-161, which yielded the best energy recovery ratio, was subjected to nutrient recovery experiments through struvite precipitation. The acid leaching method employed in this research showed high efficiencies of extraction. More

than 95 % of Mg and P in the ash were transferred to leachate. Results of precipitation experiments and nutrient balance are presented in Table 6. Approximately 3 / 4 of the nutrients in the solution was recovered as a form of a precipitate. The excess molar ratio of P compared to NH$_4$-N and Mg suggests the possible formation of phosphate precipitate other than struvite to a limited extent in association with other cations, such as calcium phosphate. The XRD patterns obtained from the precipitate matched well with the reference (PDF 01-077-2303, ICDD) from struvite (Figure 2).

If the HC-ash and PW from the selected batch of HTC are exclusively used for nutrient recovery, the molar ratio among

**Table 6. Experimental results and nutrient balance for struvite precipitation.**
PW=process water.

		[†]Precipitation experiment			[‡]Extrapolated values for full utilization of ash (proportion in total amount, %)		
		NH_4-N	Mg	P	NH_4-N	Mg	P
Input	PW	1.66	0.78	0.65	17.0 (42.4)	8.0 (20.0)	6.6 (16.5)
	Ash leachate	-	0.42	1.01	-	14.1 (35.2)	33.5 (83.5)
	Mg supplement	-	0.46	-	-	18.0 (44.8)	-
	NH_4 supplement	-	-	-	23.1 (57.6)	-	-
Total in solution		1.66	1.66	1.66	40.1	40.1	40.1
Output	Precipitate	* 1.24	1.29	1.31	30.0	31.2	31.6
	Precipitation efficiency (%)	74.7	77.7	78.9			

† Average values of duplicates, mmol

‡ Speculated values derived from the experimental results, mmol

* Elemental nitrogen obtained from CHN analysis
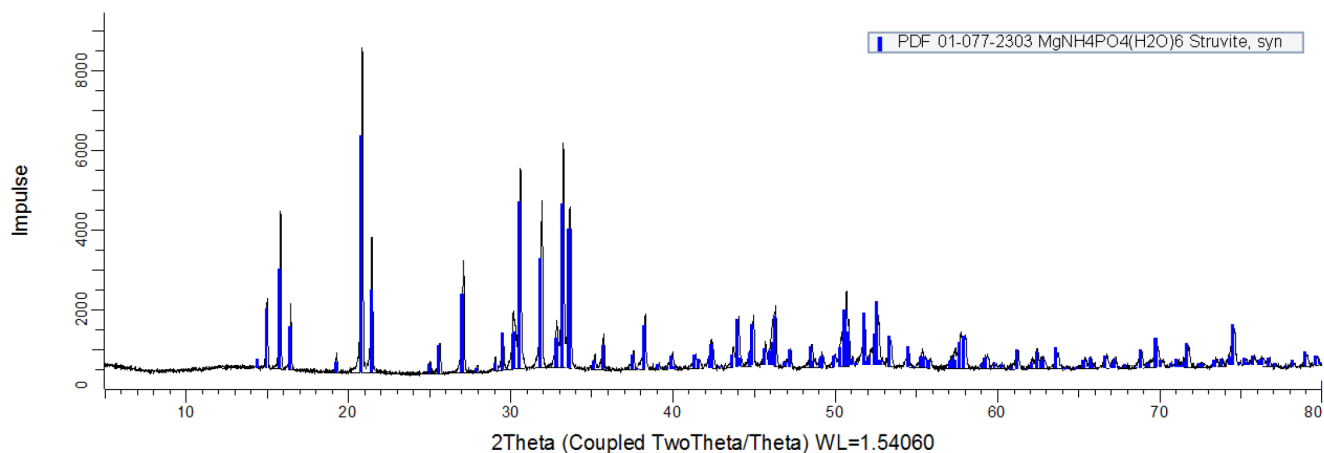
Figure 2. X-Ray diffraction analysis patterns from precipitate (Black), compared with the reference struvite (Blue).

available P: NH_4-N: Mg would be 1: 0.42: 0.55 (Table 4). It indicates that the corresponding supplements for NH_4-N and Mg are necessary to establish a stoichiometric molar ratio of 1: 1: 1 for struvite recovery. When these elements are provided, around 30 mmol of struvite, corresponding to 7.4 g $MgNH_4PO_4\cdot6H_2O$, can be harvested from hydrothermal treatment of 96 g dry faeces. After separation of the precipitate, around 1 / 4 of the total nutrient input was available in the solution at pH 8. Expected usage of acid (1M H_2SO_4) and base

(1M NaOH) in this case during the extraction-precipitation processes were 76.5 and 39.0 mL, respectively.

Discussion
Scenario for energy and nutrient recovery
This research presents a proof of concept for the hydrothermal treatment of faecal waste as an alternative sanitation technology in terms of technical feasibility, hygienization and resource recovery of energy and nutrients. The low-cost HTC reactor

with a relatively simple design and function performed well in comparison with a commercial reactor. The faecal waste was sanitized in all test conditions employed in this research, since they were greater than or comparable to the previously reported HTC conditions (at 150°C for 30 min) for complete elimination of pathogens and microbial DNA (Ducey *et al.*, 2017). The calorific values of feedstock and resulting HCs in this research correspond well with previous investigations on hydrothermal treatment of fresh human faeces (Afolabi *et al.*, 2017; Yahav Spitzer *et al.*, 2018). The HHVs of HCs ranged from 23.2 to 25.2 MJ/kg, which lies in the range of low bituminous coal, charcoal briquette and pellets produced by dry pyrolysis of human faeces. These values are greater than many of the biomass fuels currently used in the developing world (Ward *et al.*, 2014). Nutrient recovery was also demonstrated to be possible. Overall, 72 % of the phosphorus contained in the faeces could be recovered as a solid struvite, which can be used as a slow-releasing fertiliser.

Therefore, based on the tests carried out in this research, we propose a process combination for energy and nutrient recovery through hydrothermal treatment of faecal waste. The best-case scenario for resource recovery is depicted in Figure 3. In this scenario, hydrothermal treatment of 1kg dry faeces can produce 17.6 MJ and 10.3 g phosphorus in the form of solid fuel and struvite, respectively. Since the experimental results showed similar values for total energy recovery at all test conditions, the HTC condition with the lowest severity was used as the basis for the scenario. It is a more appealing option in terms of less energy consumption and ease of relevant reactor development. Furthermore, due to the limited energy potential of anaerobic digestion of PW (<6% of energy invested), we assumed that PW would be exclusively used in nutrient recovery.

Some aspects still need further investigation, such as the dewaterability of HC which can be important in the practical implementation of HTC technology by causing additional costs for separation of the solid and liquid phase of the resulting slurry. Studies have found that higher reaction severity increases the hydrophobicity of HC, which is associated with dewaterability (Sharma & Dubey, 2020; Zhang *et al.*, 2019). Though it was not quantitatively investigated in this study, observation during the experiments agrees with these reports. While the Test-161 with the lowest SF showed the best energy recovery ratio, its HC had the lowest filterability. Because we used filters with a relatively small pore size (11 μm), employing filters with a greater pore size (e.g. 63 μm) (Afolabi & Sohail, 2017) would be one of the simplest options to be tested in future investigations.

Energy use for heating the reactor can be a major operational cost factor in such a treatment system. The HCs obtained from Test-161 and BLR-161 had greater energy values than what was invested for each treatment. However, the results must be interpreted carefully and not be considered as an indication for suggesting energy independence (no external energy input needed) of treatment concept, because i) the energy input in the commercial reactor system only measured the electric energy used for heating the vessel; energy input from other units such as process parameter monitoring and controlling devices might be significant, and ii) the HHV of HC was obtained by bomb calorimetry carried out under surplus provision of oxygen. The combustion efficiency in practical use may not correspond with the laboratory results.

Also, on the laboratory scale, the energy efficiency of the system varies depending on the scale of the equipment. Water tests performed with a batch reactor system using different
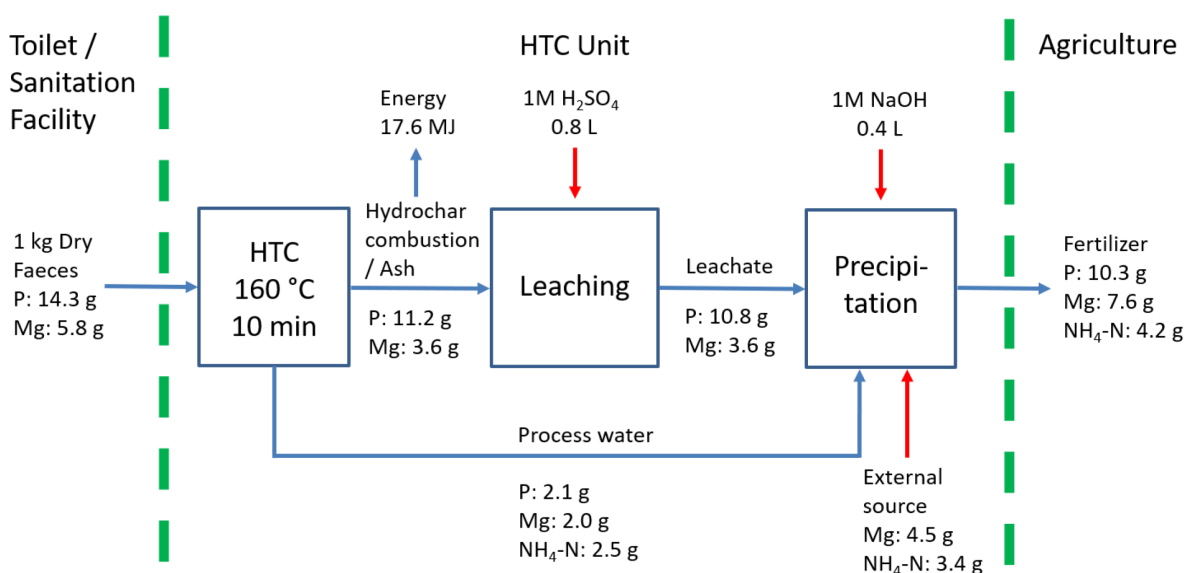

Figure 3. Scenario for energy and nutrient recovery through hydrothermal treatment of faecal waste. HTC=hydrothermal carbonization.

capacities of pressure vessels showed a significant variance in energy efficiency. The greater volume of the reactor resulted in better energy efficiency (data not shown). For a more reliable estimate of the energy demand for a real scale implementation, further research on a pilot-scale low-cost reactor system with direct energy consumption measurement is required to establish a better understanding and a model for the energy balance.

Throughout the processes, the phosphorus contained in the faeces was well retained in the system boundary. In total, 90% of the initial phosphorus in the feedstock was delivered to the precipitation unit, and finally, 72 % can be recovered in a solid phase. Because a considerable amount of nutrients were still present in the solution after separation of precipitate, further research for its reuse or enhancing the precipitation efficiency is recommended. The provision of surplus Mg over the stoichiometric ratio would be an option for better P recovery (Rahaman *et al.*, 2008). pH adjustments should also be investigated. While an increase in pH would improve the precipitation efficiency (Nelson *et al.*, 2003), an overdose of base over pH 10.5 can facilitate the formation of magnesium hydroxide compounds rather than struvite (Kim *et al.*, 2017).

Even though the production of struvite is an appealing option that can be marketed as a slow-releasing fertiliser, it may not be economical due to the cost burden from the post-processes such as acid leaching and precipitation. A more straightforward nutrient recovery option can be the direct application of resulting products from HTC. Because the HTC products may contain various chemical substances (e.g. organic acids and furan compounds) which can be beneficial or detrimental to plant growth, phytotoxicity tests with relevant conditions employing target crop and soil type need to be performed prior to their implementation in agriculture. It was reported that the increase in reaction severity resulted in greater phytotoxicity (Bargmann *et al.*, 2013). The phytotoxic compounds found in the HC in previous research were reported to be water-soluble and volatile (Bargmann *et al.*, 2013; Celletti *et al.*, 2021). Therefore, washing and/or ageing of HC can be appropriate post-treatments for decreasing the adverse impacts.

Technical feasibility of low-cost reactor concept
We developed a low-end reactor using widely available materials and operated it with a non-electric heating method. To our knowledge, this is the first attempt to use direct combustion of fuel for heating the reactor. Previous research mostly employed electric devices such as a heating mantle, microwave, oil bath or oven. This study demonstrates the feasibility of such a system and the results hold promise for the further development of small scale, decentralised and/or mobile implementations of HTC, with a possibility of being fuelled by combustion of biowastes such as crop residues or dry faecal sludge. In the current design, the amount of instrumentation was minimized in order to keep the costs down, but ensure safe operation. Of particular interest in terms of the safety assurance of low-cost reactor systems is the monitoring of the pressure increase over the heating time. Two phenomena

can affect the pressure and must be taken into account in the further reactor design and operation: i) when a closed vessel containing water and feedstock with a certain headspace is heated, pressure increases due to the saturated water vapour and gas generation from feedstock, ii) the region where the thermally expanded water fills the vessel completely, the sub-cooled liquid compression region, must be avoided. Because the pressure increases very vigorously in this region with a slight increase in temperature, the relevant experimental parameters (e.g. reactor volume, temperature, water volume and dry matter loading) should be designed carefully (Ro *et al.*, 2020).

Our observations from Test-200, 201, 206 and BLR-200 show evidence that the gas generation from feedstock can have a significant effect on the pressure increase, when it is exposed to high temperatures for a prolonged duration (Table 1 and Figure 1). Given the specifications of the current BLR design, HTC conditions more severe than BLR-200 would be inappropriate because of the rapid increase in pressure seen around 200°C. It is reassuring, however, that for all test conditions the pressure inside the reactor immediately started to decrease when the heating was stopped. It was likely that the exothermic energy from the feedstock was not sufficient for continuing the reaction (Reza *et al.*, 2014) which can induce further pressure development and cause pressure risk.

Since most of the energy input to the reactor was used for heating the reactor and water (Table 3), it may be beneficial to increase the DM loading to maximize treatment capacity and energy recovery. An important topic for future research is to investigate the interactions between the DM loading and its effects on pressure development and the properties of the resulting product.

Positioning HTC in current sanitation technologies
The HTC process can be coupled with many existing sanitation technologies due to its ability to hygienize faecal matter and to increase its fuel value. However, the specific properties of HC and PW are largely determined by the feedstock characteristics (Libra *et al.*, 2011), which can vary considerably. The characteristics of faecal sludge generated from onsite treatment facilities depends on several factors such as treatment technology, storage duration, infiltration / leakage (from the environment to system, or vice versa), collection method and climate (Strande *et al.*, 2014). The sanitation technology that best matches the approach used in this study would be the Urine-Diverting-Dry-Toilet (UDDT) which results in undiluted raw faeces that need further treatment (Riungu *et al.*, 2019). Another possible application for HTC would be as a post-treatment unit of sludge drying bed facilities, where the final product is of insufficient quality with a relatively low heating value (12 MJ/kg) and microbial risks (Seck *et al.*, 2015). Integration of hydrothermal treatment in the system can mitigate the limitations of the final product in terms of fuel quality and microbial safety assurance. The use of the BLR which could be heated by direct biomass combustion for the HTC treatment is an attractive option. The treatment could be fuelled by combusting raw dry sludge as a

low-cost fuel. In the case of faecal sludge with high moisture contents, co-HTC with other dry biowastes may be a more appealing option. Since biomass usually have a lower specific heat capacity (1.3 – 1.5 J / g dry weight at 40 °C) than water (Dupont *et al.*, 2014), its supplements will decrease the energy input for a treatment batch while producing more HC products. Also, a greater heat of reaction will be obtained from having more organic substrate in the treatment batch. A previous study reported that co-HTC of swine manure and lignocellulosic biomass showed a meaningful increase in HHV and energy content of the resulting HC materials compared to the one obtained from the treatment of pure swine manure. In addition, the concentration and bioavailability of heavy metals in HC decreased by the co-HTC (Lang *et al.*, 2018).

Conclusions

The current rate of progress in sanitation service provision in LDCs calls for breakthroughs. HTC can serve as an attractive technical solution that eliminates the microbial risk of faecal waste and converts it into valuable resources. This research provides a quantitative evaluation of the hydrothermal treatment of human waste and resource recovery. We verified the low-cost reactor concept by constructing the BLR and carrying out successive tests for faecal waste treatment. The BLR was constructed using widely available materials and technology so that local production of such hydrothermal reactors in target regions can be implemented. Also, the stable operation of the reactor using a non-electric heating method allows a wide range of heating sources to be selected. The results show that low reaction severity treatment is recommended to reduce energy consumption for treatment and increase ease of reactor construction. The resulting HC materials showed competitive heating values as solid fuel. Struvite production from HC ash and PW was demonstrated to be an efficient phosphorus recovery pathway.

Interesting future research topics include:
- phytotoxicity and plant-available nutrient analyses for faeces-derived HC, HC-ash and PW materials

- optimization of dry matter loading in HTC for enhancing energy recovery

- hydrothermal treatment of various faecal sludge samples in field conditions

- co-treatment of faecal sludge and biowaste (e.g. agricultural residue)

- pilot-scale low-cost reactor development based on locally available resources in target countries

Data availability

Underlying data

Zenodo: Underlying data / Hydrothermal carbonization as an alternative sanitation technology: process optimization and development of low-cost reactor. https://doi.org/10.5281/zenodo.5704545 (Chung *et al.*, 2021).

This project contains the following underlying data:
- HTC as an alternative sanitation technology_underlying data.xlsx

- HTC_Pressure_Temperature.xlsx

- XRD Struvite.zip

Extended data

Zenodo: Design and technical drawing of a bench-scale low-cost hydrothermal carbonization reactor. https://doi.org/10.5281/zenodo.5094908 (Chung, 2021).

This project contains the following extended data:

- 1. Design consideration.pdf

- 2. Design calculation.pdf

- 3. Technical drawing.pdf

- 4. Material list.xls

- 5. 3D visualization.stp

References

Afolabi OOD, Sohail M: **Comparative evaluation of conventional and microwave hydrothermal carbonization of human biowaste for value recovery.** *Water Sci Technol.* 2017; **75**(12): 2852–2863.
PubMed Abstract | **Publisher Full Text**

Afolabi OOD, Sohail M, Thomas CLP: **Characterization of Solid Fuel Chars recovered from Microwave Hydrothermal Carbonization of Human Biowaste.** *Energy (Oxf).* 2017; **134**: 74–89.
PubMed Abstract | **Publisher Full Text** | **Free Full Text**

Bargmann I, Rillig MC, Buss W, *et al.*: **Hydrochar and Biochar Effects on Germination of Spring Barley.** *J Agron Crop Sci.* 2013; **199**(5): 360–373.
Publisher Full Text

Bleuler M, Gold M, Strande L, *et al.*: **Pyrolysis of Dry Toilet Substrate as a Means of Nutrient Recycling in Agricultural Systems: Potential Risks and Benefits.** *Waste Biomass Valor.* 2021; **12**(7): 4171–4183.
Publisher Full Text

Boot NL, Scott RE: **Faecal sludge in Accra, Ghana: problems of urban

provision. *Water Sci Technol.* 2009; **60**(3): 623–631.
PubMed Abstract | **Publisher Full Text**

Celletti S, Bergamo A, Benedetti V, *et al.*: **Phytotoxicity of hydrochars obtained by hydrothermal carbonization of manure-based digestate.** *J Environ Manage.* 2021; **280**: 111635.
PubMed Abstract | **Publisher Full Text**

Chung JW, Ovsyannikova E, Treichler A, *et al.*: **Underlying data / Hydrothermal carbonization as an alternative sanitation technology: process optimization and development of low-cost reactor.** *Zenodo.* 2021.
http://www.doi.org/10.5281/zenodo.5704545

Chung JW: **Design and technical drawing of a bench-scale low-cost hydrothermal carbonization reactor.** *Zenodo.* 2021.
http://www.doi.org/10.5281/zenodo.5094908

Cordell D, Drangert JO, White S: **The story of phosphorus: Global food security and food for thought.** *Glob Environ Change.* 2009; **19**(2): 292–305.
Publisher Full Text

Cordell D, Rosemarin A, Schröder JJ, *et al.*: **Towards global phosphorus security: A systems framework for phosphorus recovery and reuse options.** *Chemosphere.* 2011; **84**(6): 747–758.
PubMed Abstract | **Publisher Full Text**

Danso-Boateng E, Holdich RG, Martin SJ, *et al.*: **Process energetics for the hydrothermal carbonisation of human faecal wastes.** *Energy Convers Manag.* 2015; **105**: 1115–1124.
Publisher Full Text

Danso-Boateng E, Holdich RG, Shama G, *et al.*: **Kinetics of faecal biomass hydrothermal carbonisation for hydrochar production.** *Appl Energy.* 2013; **111**: 351–357.
Publisher Full Text

Desmidt E, Ghyselbrecht K, Zhang Y, *et al.*: **Global Phosphorus Scarcity and Full-Scale P-Recovery Techniques: A Review.** *Crit Rev Environ Sci Technol.* 2015; **45**(4): 336–384.
Publisher Full Text

Ducey TF, Collins JC, Ro KS, *et al.*: **Hydrothermal carbonization of livestock mortality for the reduction of pathogens and microbially-derived DNA.** *Front Environ Sci Eng.* 2017; **11**(3): 9.
Publisher Full Text

Dupont C, Chiriac R, Gauthier G, *et al.*: **Heat capacity measurements of various biomass types and pyrolysis residues.** *Fuel.* 2014; **115**: 644–651.
Publisher Full Text

Fakkaew K, Koottatep T, Polprasert C: **Faecal sludge treatment and utilization by hydrothermal carbonization.** *J Environ Manage.* 2018; **216**: 421–426.
PubMed Abstract | **Publisher Full Text**

Heidari M, Norouzi O, Salaudeen S, *et al.*: **Prediction of Hydrothermal Carbonization with Respect to the Biomass Components and Severity Factor.** *Energy & Fuels.* 2019; **33**(10): 9916–9924.
Publisher Full Text

Kim D, Min KJ, Lee K, *et al.*: **Effects of pH, molar ratios and pre-treatment on phosphorus recovery through struvite crystallization from effluent of anaerobically digested swine wastewater.** *Environ Eng Res.* 2017; **22**(1): 12–18.
Reference Source

Lang Q, Guo Y, Zheng Q, *et al.*: **Co-hydrothermal carbonization of lignocellulosic biomass and swine manure: Hydrochar properties and heavy metal transformation behavior.** *Bioresour Technol.* 2018; **266**: 242–248.
PubMed Abstract | **Publisher Full Text**

Libra JA, Ro KS, Kammann C, *et al.*: **Hydrothermal carbonization of biomass residuals: a comparative review of the chemistry, processes and applications of wet and dry pyrolysis.** *Biofuels.* 2011; **2**(1): 71–106.
Publisher Full Text

Mallory A, Holm R, Parker A: **A Review of the Financial Value of Faecal Sludge Reuse in Low-Income Countries.** *Sustainability.* 2020; **12**(20): 8334.
Publisher Full Text

Namioka T, Miyazaki M, Morohashi Y, *et al.*: **Modeling and Analysis of Batch-Type Thermal Sludge Pretreatment for Optimal Design.** *Journal of Environment and Engineering.* 2008; **3**(1): 170–181.
Publisher Full Text

Nelson NO, Mikkelsen RL, Hesterberg DL: **Struvite precipitation in anaerobic swine lagoon liquid: effect of pH and Mg:P ratio and determination of rate constant.** *Bioresour Technol.* 2003; **89**(3): 229–236.
PubMed Abstract | **Publisher Full Text**

Ovsyannikova E, Kruse A, Becker GC: **Feedstock-Dependent Phosphate Recovery in a Pilot-Scale Hydrothermal Liquefaction Bio-Crude Production.**

Energies. 2020; **13**(2): 379.
Publisher Full Text

Ovsyannikova E, Kruse A, Becker GC: **Valorization of Byproducts from Hydrothermal Liquefaction of Sewage Sludge and Manure: the Development of a Struvite-Producing Unit for Nutrient Recovery.** *Energy & Fuels.* 2021; **35**(11): 9408–9423.
Publisher Full Text

Petzet S, Peplinski B, Cornel P: **On wet chemical phosphorus recovery from sewage sludge ash by acidic or alkaline leaching and an optimized combination of both.** *Water Res.* 2012; **46**(12): 3769–3780.
PubMed Abstract | **Publisher Full Text**

Rahaman MS, Ellis N, Mavinic DS: **Effects of various process parameters on struvite precipitation kinetics and subsequent determination of rate constants.** *Water Sci Technol.* 2008; **57**(5): 647–654.
PubMed Abstract | **Publisher Full Text**

Reza MT, Andert J, Wirth B, *et al.*: **Hydrothermal Carbonization of Biomass for Energy and Crop Production.** *Applied Bioenergy.* 2014; **1**(1): 11–29.
Publisher Full Text

Riungu J, Ronteltap M, van Lier JB: **Anaerobic stabilisation of urine diverting dehydrating toilet faeces (UDDT-F) in urban poor settlements: biochemical energy recovery.** *Journal of Water, Sanitation and Hygiene for Development.* 2019; **9**(2): 289–299.
Publisher Full Text

Ro KS, Libra JA, Alvarez-Murillo A: **Comparative Studies on Water- and Vapor-Based Hydrothermal Carbonization: Process Analysis.** *Energies.* 2020; **13**(21): 5733.
Publisher Full Text

Robbiani Z: **Hydrothermal carbonization of biowaste/fecal sludge**. Master's thesis, Swiss Federal Institute of Technology Zurich, 2013.
Reference Source

Seck A, Gold M, Niang S, *et al.*: **Faecal sludge drying beds: increasing drying rates for fuel resource recovery in Sub-Saharan Africa.** *J Water Sanit Hyg Dev.* 2015; **5**(1): 72–80.
Publisher Full Text

Sharma HB, Dubey BK: **Binderless fuel pellets from hydrothermal carbonization of municipal yard waste: Effect of severity factor on the hydrochar pellets properties.** *J Clean Prod.* 2020; **277**: 124295.
Publisher Full Text

Sharma R, Jasrotia K, Singh N, *et al.*: **A Comprehensive Review on Hydrothermal Carbonization of Biomass and its Applications.** *Chemistry Africa.* 2020; **3**(1): 1–19.
Publisher Full Text

Song C, Zheng H, Shan S, *et al.*: **Low-Temperature Hydrothermal Carbonization of Fresh Pig Manure: Effects of Temperature on Characteristics of Hydrochars.** *J Environ Engineer.* 2019; **145**(6): 04019029.
Publisher Full Text

Strande L, Ronteltap M, Brdjanovic D: **Faecal Sludge Management: Systems Approach for Implementation and Operation.** IWA Publishing, 2014; **13**.
Publisher Full Text

VDI: **Fermentation of organic materials - Characterisation of the substrate, sampling, collection of material data, fermentation tests.** Verein Deutscher Ingenieure Düsseldorf, Germany, 2006.
Reference Source

Ward BJ, Yacob TW, Montoya LD: **Evaluation of Solid Fuel Char Briquettes from Human Waste.** *Environ Sci Technol.* 2014; **48**(16): 9852–9858.
PubMed Abstract | **Publisher Full Text**

WHO, UNICEF: **Progress on household drinking water, sanitation and hygiene 2000-2020: five years into the SDGs.** Geneva: World Health Organization (WHO) and the United Nations Children's Fund (UNICEF), Licence: CC BY-NC-SA 3.0 IGO, 2021.
Reference Source

Wüst D, Rodriguez Correa C, Suwelack KU, *et al.*: **Hydrothermal carbonization of dry toilet residues as an added-value strategy - Investigation of process parameters.** *J Environ Manage.* 2019; **234**: 537–545.
PubMed Abstract | **Publisher Full Text**

Yahav Spitzer R, Mau V, Gross A: **Using hydrothermal carbonization for sustainable treatment and reuse of human excreta.** *J Clean Prod.* 2018; **205**: 955–963.
Publisher Full Text

Zhang S, Zhu X, Zhou S, *et al.*: **Biochar from Biomass and Waste.** Ok, Y.S., Tsang, D.C.W., Bolan, N. and Novak, J.M. (eds), Elsevier, 2019; 275–294.
Publisher Full Text

Open Peer Review

Current Peer Review Status: ❓ ❓

Version 1

Reviewer Report 18 January 2022

https://doi.org/10.21956/openreseurope.15435.r28090

❓ **Amit Gross** [iD]

Zuckerberg Institute for Water Research, The Jacob Blaustein Institutes for Desert Research, Ben-Gurion University of the Negev, Be'er Sheva, Israel

I appreciate your work, I have enjoyed reading it. Overall I think it is a good paper with scientific merit to HTC of human feces. I do have some fairly minor comments and questions.

Method section:
  ○ I could not understand the decision to sort different components because it does not represent correctly human excretions nor the overall waste from a composting toilet. Thus, not really represent any real feedstock. Can you comment on the reasoning?

  ○ Statistical analysis is somewhat weak, specifically as you used only duplicates.
Results Section:
  ○ For clarity please revise Figure 1 legends.
Discussion section
  ○ Please add a discussion about the quality of the char for combustion.

Is the work clearly and accurately presented and does it cite the current literature?
Yes

Is the study design appropriate and does the work have academic merit?
Partly

Are sufficient details of methods and analysis provided to allow replication by others?
Yes

If applicable, is the statistical analysis and its interpretation appropriate?
Partly

Are all the source data underlying the results available to ensure full reproducibility?

Yes

Are the conclusions drawn adequately supported by the results?
Partly

Competing Interests: No competing interests were disclosed.

Reviewer Expertise: Currently study HTC of organic waste including human excretion.

I confirm that I have read this submission and believe that I have an appropriate level of expertise to confirm that it is of an acceptable scientific standard, however I have significant reservations, as outlined above.

> Author Response 24 May 2022
> **Jae Wook Chung**, Zurich University of Applied Sciences, Wädenswil, Switzerland
>
> Dear Reviewers, We appreciate your comments. They were beneficial in improving this article, making it more precise and comprehensive. We accepted most of your comments and revised the manuscript accordingly. Please find our responses to the comments for the cases we would like to maintain the original manuscript or need further clarification. We hope the second version of the manuscript addresses your comments sufficiently.
>
> *Reviewer 2 / comments I appreciate your work, I have enjoyed reading it. Overall I think it is a good paper with scientific merit to HTC of human feces. I do have some fairly minor comments and questions.*
>
> *Method section:*
> - *I could not understand the decision to sort different components because it does not represent correctly human excretions nor the overall waste from a composting toilet. Thus, not really represent any real feedstock. Can you comment on the reasoning?*
>
> **Our response:** We agree that the faecal stock used in the HTC experiments does not represent any raw faecal sludges in field conditions. Pre-treatments of faeces were unavoidable to meet the biosafety requirement in laboratory research. This aspect is described in the Methods and Discussion section of the revised manuscript as recommended by Reviewer 1. As described in the discussion section, the property of faecal sludge varies greatly depending on many factors. Thus, we selected pure human waste as the main component of various faecal sludges, especially ones obtained from Urine Diverting Dry Toilet or box sanitation. As an alternative to sorting out faecal mass, we tested autoclaving-drying-grinding of faecal stocks as received (faeces + toilet paper + woody cover materials). However, it was challenging to prepare homogeneous feedstock because the high fibre content of large paper materials interrupted the mechanical grinding process. The most reliable and necessary investigation would be testing the field samples in LDC conditions. It is suggested as further research topics.
> - *Statistical analysis is somewhat weak, specifically as you used only duplicates.*
>
> **Our response:** Performing at least triplicate of each experimental condition would have provided a more robust statistical basis. However, one of the main difficulties of this

research was collecting enough undiluted human faeces. Based on the minor differences observed from the duplicate of each experimental condition (Table 1), we considered that the reported results are reliable.

Results Section:
- *For clarity please revise Figure 1 legends.*

Our response: Figure 1 and its legends are revised.

Discussion section
- *Please add a discussion about the quality of the char for combustion.*

Our response: Relevant information is provided in the discussion section.

Competing Interests: No competing interests were disclosed.

Reviewer Report 23 December 2021

https://doi.org/10.21956/openreseurope.15435.r28088

? **Luca Fiori** (iD)

Department of Civil, Environmental and Mechanical Engineering & C3A - Center Agriculture Food Environment, University of Trento, Trento, Italy

Donato Scrinzi (iD)

Department of Civil, Environmental and Mechanical Engineering, University of Trento, Trento, Italy

The research article "Hydrothermal carbonization as an alternative sanitation technology: process optimization and development of low-cost reactor" is well written, and the research is meaningful and original. The authors cover many interesting aspects of HTC applied in a developing countries context for sanitation and struvite production from human wastes. It is definitely an interesting work, and we suggest it should be indexed after revision. The points here reported could help in that.

INTRODUCTION.
Just a couple of suggestions.
1. "applying higher temperatures and longer retention times>." We suggest replacing "retention times" with "reaction times".

2. "A batch type reactor with 20 L capacity was constructed employing a relatively simple design and function at a material cost of around CHF 11K." We suggest indicating costs in euros (or U.S. dollars) – at least in brackets.

MATERIALS AND METHODS.
Human waste feedstock and hydrochar.
1. The feedstocks used for HTC process were two stocks of faeces, which were dried and then re-moisturized to 20% dry matter. We wonder how much the initial moisture content in the untreated faeces is. The approach used by the authors is understandable and reasonable in a laboratory research context. However, the authors must mention that in the context of the practical realization of the concept, the raw material would be treated as it is, without preliminary drying. Having said that, as a matter of fact, the drying process may change some properties (like NH4-N content, dewaterability), thus the final properties of HTC products. We suggest mentioning this in the paper.

2. We suggest the authors explicitly write the extended form "relative centrifugal force" before the acronym RCF.

Process optimization in commercial batch reactor
1. The authors could consider explaining already in this part which are the tested HTC process temperatures and durations, after "several experimental conditions were tested."

2. Moreover, the idea to test Touch & Down (TD) condition is very smart for low-cost applications without a heating controller, especially in the context of developing countries. Is there a particular reason for testing TD condition only for 200 °C and not for the other lower temperatures?

3. The naming for samples is not so straightforward. As a suggestion, it might be made clearer writing in full the Celsius degrees for reaction temperature and decades of minutes for duration, with TD considered as 0 minutes, e.g. instead of << Test-161 for HTC at 160 °C for 10 minutes and Test-206 for HTC at 200 °C for 60 minutes. The TD case, where the heating stopped when TR = 200 °C, is named Test-200.>> writing Test-160-10, Test 200-60, and Test-200-0, respectively.

4. We think the introduction of SF_adj is smart to make the severity factor more adherent to the real conditions and comparable. However, we found it difficult to understand the difference between SF vs SF_adj at a first reading. If we understood correctly reading Table 1, t_adj is defined as the time interval in which the reactor actually stays inside the range of ±3% of for the set temperature (and not a tolerance for time as I initially thought). We would suggest rephrasing the definition of t_adj in order to avoid any misconception.

Material characterization
1. Since it is crucial in the experimental procedure for ash production, the authors should specify at what temperature and residence time the muffle furnace worked for ash content analysis (for readers who do not have access to the standard DIN EN 14775). The same applies also for VS analyses (2540-E).

Energy analysis
1. There is some information useful for reproducibility about BMP tests which is missing. In particular, we suggest the authors expand this section with the substrate to inoculum ratio (SIR, referred to VS or COD) resulting from the mass chosen.

2. The use of thymolphthalein as pH indicator is interesting and it seems widely used in literature. As far as we can understand, this indicator can be used as a color signal for solution replacement (uncolored when pH < 9.3-10.5), in order to guarantee a proper CO_2

removal. For the sake of reproducibility, the authors may add a reference or explain better how it can be useful. We found this research interesting (https://doi.org/10.1002/tqem.21776) even if not fully explicative, but hopefully the authors may have better references to mention.

3. Before the process energy input for BLR tests, we suggest the authors mention how the energy input for tests with commercial reactor is considered/calculated (since it is shown in Table 2), and not only in the discussion.

4. Mg0 could be better explained as *fuel* gas bottle (since 'gas' can be confused with the aforementioned gas produced by BMP bottles, which could be mentioned as 'biogas' to avoid any misunderstanding).

5. "HV is the heating value of gas": specify HHV or LHV.

6. "H_T is the enthalpy of saturated water at T." In eq. (4) it is reported as H_r: modify accordingly.

7. Consider to further explain the calculation of energy content of the HTC products (as far as we understand, it seems HHV_HC*mass_HC for solids and HHV_methane*mass_biomethane produced for process water).

RESULTS
HTC of faeces
1. "Much more pressure was produced in BLR-200 than in Test-200." We suggest to rephrase: the gas is produced, not the pressure.

2. "and approached the maximum operational pressure." Specify here in the main text that the pressure limit for BLR is 25 bar (data available in Extended Data)

3. We suggest to improve the quality of Figure 1: in terms of both graphics and legend: it is quite hard to follow the various curves in association with their relevant variables. Try to make it more immediate to read and understand.

4. Table 1 could be shown in the following page, closer to the results section.

Energy contents
1. Check the orthography: kJ, not KJ in capital letter (also in Table 2).

2. Table 2 (but valid in general), please explicitly mention in the units if the mass considered is on a dry basis or as received. The unit of total energy recovery ratio should be indicated as (%).

3. Total energy recovery ratio could be compared also to the energy that was present inside the initial feedstock (Table 2).

4. BMP results are slightly mentioned in the text and not compared with other works.

5. "The exothermic energy from the hydrothermal reaction of feedstock contributed to the total energy demand to a limited extent (< 4% of energy input)." Recent literature specifically

dedicated to the evaluation of the exothermicity of the HTC reaction further corroborates this finding.

Nutrient contents

1. Table 1. Something is not right with respect to the data in table 1. The authors states that the "Oxygen contents estimated by subtracting the CHNS and ash fraction from 100 %." But if we sum the CHNSO and ASH % we get values other than 100, beyond issues that can be related to rounding (e.g. for Faeces-1 we get 98.4). Check the correctness of all the data in Table 1.

2. Table 3. As a suggestion, the caption could be changed from 'energy inputs' to 'Energy assessment'. Moreover, there must be an agreement between the convention on signs for endo- and exothermal reactions in both materials and method and results sections, e.g., changing the definition of equations (3) and (4) as $\Sigma_i(Energy)_i = 0$ (all terms are positive if given to the system and negative if released by the system).

3. At the beginning of pg. 9, Table 5 can be explicitly named when referring to data of metals.

4. Table 4. 'Loss from HTC' does not seem the correct name, we'd rather say 'Loss from experimental procedure/Loss after HTC'. The P mass balance should be closed (P mass is conservative).

5. The calculations for partitioning ratio in HC and PW are correct, but they can be explained in more detail in this section or, better, in the materials and methods section.

Struvite precipitation

1. The difference in the ratio among P:NH$_4$:Mg could not be exactly the same for other phosphates precipitation, as indicated by the authors, but there could be also a not-statistically-significant difference inside the experimental error of the measurements (here missing).

DISCUSSION

Scenario for energy and nutrient recovery

1. Figure 3. Not all output streams are indicated (solid residues from leaching, liquid stream after struvite precipitation, etc). It can be useful to explicitly show them for further studies on recovery and environmental assessments of output waste streams. Moreover, for completeness' sake, NH$_4$-N initial value could be indicated also in the toilet/sanitation facility.

Other general considerations.

We like the open approach by the journal and the authors' decision to make available Extended Data with all the design detail of the reactor. Beyond the need to revise some aspects of the paper as highlighted above, the paper is of scientific interest and frames very well the problem of the management of human faeces in developing contexts that the authors propose to solve with HTC technology with basically simple reactors. Beyond the interesting experimental results, the authors analyse and discuss the advantages and limitations of the technology in the application context, from nutrient recovery to energy recovery.

As a whole, a good paper that, after careful review by the authors, will deserve to be indexed.

References

1. Konrad O, Hasan C, Marder M, Zulian L, et al.: Comparison of two gas volume measurement

systems by evaluating biochemical methane potential. *Environmental Quality Management*. 2021.
Publisher Full Text

Is the work clearly and accurately presented and does it cite the current literature?
Partly

Is the study design appropriate and does the work have academic merit?
Yes

Are sufficient details of methods and analysis provided to allow replication by others?
Partly

If applicable, is the statistical analysis and its interpretation appropriate?
I cannot comment. A qualified statistician is required.

Are all the source data underlying the results available to ensure full reproducibility?
Yes

Are the conclusions drawn adequately supported by the results?
Yes

Competing Interests: No competing interests were disclosed.

Reviewer Expertise: We (Luca Fiori and Donato Scrinzi) feel qualified to review and assess all the aspects of the paper.

We confirm that we have read this submission and believe that we have an appropriate level of expertise to confirm that it is of an acceptable scientific standard, however we have significant reservations, as outlined above.

Author Response 24 May 2022
Jae Wook Chung, Zurich University of Applied Sciences, Wädenswil, Switzerland

Dear Reviewers, We appreciate your comments. They were beneficial in improving this article, making it more precise and comprehensive. We accepted most of your comments and revised the manuscript accordingly. Please find our responses to the comments. We hope the second version of the manuscript addresses your comments sufficiently.

Reviewer 1 / Comments
The research article "Hydrothermal carbonization as an alternative sanitation technology: process optimization and development of low-cost reactor" is well written, and the research is meaningful and original. The authors cover many interesting aspects of HTC applied in a developing countries context for sanitation and struvite production from human wastes. It is definitely an interesting work, and we suggest it should be indexed after revision. The points here reported could help in that.

INTRODUCTION.

Just a couple of suggestions.

"applying higher temperatures and longer retention times>." We suggest replacing "retention times" with "reaction times".

Our response: The manuscript is revised accordingly (Introduction, paragraph 3)

"A batch type reactor with 20 L capacity was constructed employing a relatively simple design and function at a material cost of around CHF 11K." We suggest indicating costs in euros (or U.S. dollars) – at least in brackets.

Our response: The cost is given in EUR (Introduction, paragraph 5).

MATERIALS AND METHODS.

Human waste feedstock and hydrochar.

The feedstocks used for HTC process were two stocks of faeces, which were dried and then re-moisturized to 20% dry matter. We wonder how much the initial moisture content in the untreated faeces is. The approach used by the authors is understandable and reasonable in a laboratory research context. However, the authors must mention that in the context of the practical realization of the concept, the raw material would be treated as it is, without preliminary drying. Having said that, as a matter of fact, the drying process may change some properties (like NH4-N content, dewaterability), thus the final properties of HTC products. We suggest mentioning this in the paper.

Our response: The primary consideration of the human waste experiments was assuring biosafety. Thus, all faecal stocks were autoclaved before introducing them into lab facilities. To provide a more explicit justification of the concept and limitation of this research, we provided more information on the feedstock and its preparation in the Methods section (Human waste feedstock and hydrochar). Also, we added the possible impact of feedstock pre-treatment in the discussion part (Positioning HTC in current sanitation technologies).

We suggest the authors explicitly write the extended form "relative centrifugal force" before the acronym RCF.

Our response: The manuscript is revised accordingly.

Process optimization in commercial batch reactor

The authors could consider explaining already in this part which are the tested HTC process temperatures and durations, after "several experimental conditions were tested."

Our response: The manuscript is revised accordingly.

Moreover, the idea to test Touch & Down (TD) condition is very smart for low-cost applications without a heating controller, especially in the context of developing countries. Is there a particular reason for testing TD condition only for 200 °C and not for the other lower temperatures?

Our response: Testing more TD conditions in a lower temperature range would have provided valuable information for test conditions. However, it was limited by the amount of available feedstock, which was one of the main practical limitations. Based on the results from optimisation experiments, we expected that TD treatment in a lower temperature range (160-180 °C) would result in insufficient dewaterability of the resulting slurry from HTC treatment. The relevant information is provided in the Methods section (Process

optimization in commercial batch reactor).

The naming for samples is not so straightforward. As a suggestion, it might be made clearer writing in full the Celsius degrees for reaction temperature and decades of minutes for duration, with TD considered as 0 minutes, e.g. instead of << Test-161 for HTC at 160 °C for 10 minutes and Test-206 for HTC at 200 °C for 60 minutes. The TD case, where the heating stopped when TR = 200 °C, is named Test-200.>> writing Test-160-10, Test 200-60, and Test-200-0, respectively.
Our response: The sample and Test IDs are revised as recommended throughout the manuscript.

We think the introduction of SF_adj is smart to make the severity factor more adherent to the real conditions and comparable. However, we found it difficult to understand the difference between SF vs SF_adj at a first reading. If we understood correctly reading Table 1, t_adj is defined as the time interval in which the reactor actually stays inside the range of ±3% of for the set temperature (and not a tolerance for time as I initially thought). We would suggest rephrasing the definition of t_adj in order to avoid any misconception.
Our response: The manuscript is revised accordingly.

Material characterization
Since it is crucial in the experimental procedure for ash production, the authors should specify at what temperature and residence time the muffle furnace worked for ash content analysis (for readers who do not have access to the standard DIN EN 14775). The same applies also for VS analyses (2540-E).
Our response: The relevant test conditions are provided.

Energy analysis
There is some information useful for reproducibility about BMP tests which is missing. In particular, we suggest the authors expand this section with the substrate to inoculum ratio (SIR, referred to VS or COD) resulting from the mass chosen.
Our response: VS-substrate (VSS) to VS-inoculum (VSI) ratio is provided.

The use of thymolphthalein as pH indicator is interesting and it seems widely used in literature. As far as we can understand, this indicator can be used as a color signal for solution replacement (uncolored when pH < 9.3-10.5), in order to guarantee a proper CO_2 removal. For the sake of reproducibility, the authors may add a reference or explain better how it can be useful. We found this research interesting (https://doi.org/10.1002/tqem.21776) even if not fully explicative, but hopefully the authors may have better references to mention. (Konrad et al., 2021)
Our response: We think that the use of thymolphthalein as a pH indicator is quite common practice in biogas laboratories. The relevant information is provided with the suggested reference.

Before the process energy input for BLR tests, we suggest the authors mention how the energy input for tests with commercial reactor is considered/calculated (since it is shown in Table 2), and not only in the discussion.
Our response: The relevant information is provided (at the end of Energy analysis section)

Mg0 could be better explained as fuel gas bottle (since 'gas' can be confused with the

aforementioned gas produced by BMP bottles, which could be mentioned as 'biogas' to avoid any misunderstanding)
Our response: The manuscript is revised accordingly.

"HV is the heating value of gas": specify HHV or LHV.
Our response: It is HHV. The manuscript is revised.

"H_T is the enthalpy of saturated water at T." In eq. (4) it is reported as H_r: modify accordingly.
Our response: The manuscript is revised accordingly.

Consider to further explain the calculation of energy content of the HTC products (as far as we understand, it seems HHV_HC*mass_HC for solids and HHV_methane*mass_biomethane produced for process water).
Our response: The manuscript is revised accordingly.

RESULTS
HTC of faeces
"Much more pressure was produced in BLR-200 than in Test-200." We suggest to rephrase: the gas is produced, not the pressure.
Our response: The manuscript is revised accordingly.

"and approached the maximum operational pressure." Specify here in the main text that the pressure limit for BLR is 25 bar (data available in Extended Data)
Our response: The relevant information is provided both in the Methods (Bench-scale low-cost reactor) and Results section (HTC of faeces, paragraph 3).

We suggest to improve the quality of Figure 1: in terms of both graphics and legend: it is quite hard to follow the various curves in association with their relevant variables. Try to make it more immediate to read and understand.
Our response: The graph and legend of Figure 1 are revised.

Table 1 could be shown in the following page, closer to the results section.
Our response: It is noted to the editorial team.

Energy contents
Check the orthography: kJ, not KJ in capital letter (also in Table 2)
Our response: The manuscript is revised accordingly.

Table 2 (but valid in general), please explicitly mention in the units if the mass considered is on a dry basis or as received. The unit of total energy recovery ratio should be indicated as (%).
Our response: The manuscript is revised accordingly.

Total energy recovery ratio could be compared also to the energy that was present inside the initial feedstock (Table 2). **Our response:** *The manuscript is revised accordingly (Results - Energy contents section and Table 2).*

BMP results are slightly mentioned in the text and not compared with other works.

Our response: The relevant information is added to the Discussion as a new paragraph. Also, the first part of the Discussion (Scenario for energy and nutrient recovery) is re-structured: an overview of the HTC process and resource recovery efficiency is provided. Then, energy recovery through solid fuel and biogas production is discussed in detail.

"The exothermic energy from the hydrothermal reaction of feedstock contributed to the total energy demand to a limited extent (< 4% of energy input)." Recent literature specifically dedicated to the evaluation of the exothermicity of the HTC reaction further corroborates this finding
Our response: We added the corresponding information to the Discussion part (Technical feasibility of low-cost reactor concept) with a reference (Ischia *et al.*, 2022).

Nutrient contents
Table 1. Something is not right with respect to the data in table 1. The authors states that the "Oxygen contents estimated by subtracting the CHNS and ash fraction from 100 %." But if we sum the CHNSO and ASH % we get values other than 100, beyond issues that can be related to rounding (e.g. for Faeces-1 we get 98.4). Check the correctness of all the data in Table 1.
Our response: The contents of Table 1 are corrected.

Table 3. As a suggestion, the caption could be changed from 'energy inputs' to 'Energy assessment'. Moreover, there must be an agreement between the convention on signs for endo- and exothermal reactions in both materials and method and results sections, e.g., changing the definition of equations (3) and (4) as $\Sigma i(Energy)i=0$ (all terms are positive if given to the system and negative if released by the system).
Our response: The caption of Table 3 is revised as recommended. The signs of the energy terms now follow the convention of thermodynamics in both the material and methods and Table 3.

At the beginning of pg. 9, Table 5 can be explicitly named when referring to data of metals.
Our response: We would like to maintain the original manuscript. We consider it is more appropriate to present the data as nutrients because they also include non-metallic substances: P and NH4-N.

Table 4. 'Loss from HTC' does not seem the correct name, we'd rather say 'Loss from experimental procedure/Loss after HTC'. The P mass balance should be closed (P mass is conservative).
Our response: The manuscript is revised accordingly.

The calculations for partitioning ratio in HC and PW are correct, but they can be explained in more detail in this section or, better, in the materials and methods section.
Our response: The relevant information is added in the Materials section (Material characterization).

Struvite precipitation
The difference in the ratio among P:NH$_4$:Mg could not be exactly the same for other phosphates precipitation, as indicated by the authors, but there could be also a not-statistically-significant difference inside the experimental error of the measurements (here missing).
Our response: We agree that there might be insignificant experimental errors. However, we do not have enough tests to clear statistical insignificance. Instead, we think that the

different analytical methods would result in the imbalance of P:NH4:Mg ratio. The manuscript is revised by adding this explanation.

DISCUSSION
Scenario for energy and nutrient recovery

Figure 3. Not all output streams are indicated (solid residues from leaching, liquid stream after struvite precipitation, etc). It can be useful to explicitly show them for further studies on recovery and environmental assessments of output waste streams. Moreover, for completeness' sake, NH_4-N initial value could be indicated also in the toilet/sanitation facility.

Our response: We revised Figure 3 by adding nutrient contents: remaining amounts in the solid residue from acid leaching and out-solution from the precipitation process. However, NH_4-N contents of solid samples were out of our research scope. Nutrient recovery was only considered after the combustion of hydrochar, and we assumed that ammonium content in HC was completely volatilised during the combustion of HC. Figure 3 and its caption are revised by providing more comments on NH4-N contents.

Competing Interests: No competing interests were disclosed.